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1 **Title: Anatectic melt inclusions in ultra-high temperature granulites**

2 **Omar Gianola**¹, Omar Bartoli¹, Bernardo Cesare¹, Fabio Ferri¹, Andrea Galli², Silvio Ferrero³,
3 Luca Capizzi^{1,4}, Christian Liebske², Laurent Remusat⁵, Stefano Poli⁶

4 ¹ Dipartimento di Geoscienze, Università degli Studi di Padova, Via G. Gradenigo 6, 35131 Padova,
5 Italy

6 ² Institut für Geochemie und Petrologie, ETH Zürich, Switzerland

7 ³ Institut für Erd- und Umweltwissenschaften, Universität Potsdam, Potsdam, Germany

8 ⁴ Dipartimento di Scienze della Terra, Università degli Studi di Roma “La Sapienza”, Roma, Italy

9 ⁵ Institut de Minéralogie, de Physique des Matériaux, et de Cosmochimie (IMPMC), UMR CNRS 7590
10 – Sorbonne Université – Muséum National d'Histoire Naturelle, Paris, France

11 ⁶ Dipartimento di Scienze della Terra “Ardito Desio”, Università degli Studi di Milano, Milano, Italy

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13 E-mail: omar.gianola@unipd.it

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15 **Key words:** Fluid inclusions; Garnet; Granites; Gruf Complex; Nanogranitoids

16

17 **Abstract**

18 Partial melting up to ultra-high temperature (UHT) conditions is one of the major processes for
19 the geochemical differentiation and reworking of the mid- to lower continental crust, with
20 relevant implications on its rheological behaviour. UHT granulites from the Gruf Complex
21 (Central Alps) display garnet and sapphirine porphyroblasts containing a variety of primary
22 melt inclusions. Typically, melt inclusions in garnets occur as glassy and polycrystalline

23 inclusions (i.e. nanogranitoids), the latter often organized in mm-sized clusters associated with
24 primary fluid inclusions. Nanogranitoids are characterized by an elliptical faceted shape, with
25 variable sizes ranging from 2 to 115 μm , while glassy inclusions show negative crystal shapes
26 that usually never exceed 15 μm in diameter and present CO_2 -rich shrinkage bubbles. The
27 characteristic mineral assemblage observed in nanogranitoids consists of quartz, biotite,
28 muscovite, plagioclase, K-feldspar, kokchetavite and very rarely aluminosilicates. Glassy and
29 re-homogenized melt inclusions are peraluminous and rhyolitic in composition, with $\text{SiO}_2 = 69$
30 $- 80$ wt% and $\text{Na}_2\text{O} + \text{K}_2\text{O} = 5 - 12$ wt%. Commonly, the analysed melt inclusions have very
31 high K_2O (> 6 wt%) and very low Na_2O (< 2 wt%) contents, indicative for potassic to
32 ultrapotassic melts. Measured H_2O contents of the melts range from 2.9 to 8.8 wt%, whereas
33 CO_2 concentrations are between 160 and 1738 ppm. Accordingly, calculated viscosities for re-
34 homogenized melt inclusions vary between 10^4 and 10^5 Pa·s. Related primary fluid inclusions
35 mainly contain CO_2 , with rare occurrence of CO and N_2 , and are often associated with quartz,
36 as well as different carbonates and phyllosilicates. It is assumed that the source for the carbonic
37 fluid was external and probably related to the degassing lithospheric mantle. Consequently, it
38 is argued that anatexis was initially triggered by incongruent dehydration melting reactions
39 involving biotite breakdown and proceeded in the presence of an externally derived COH-
40 bearing fluid. The coexistence of COH-bearing fluid and melt inclusions indicates that partial
41 melting occurred under conditions of fluid-melt immiscibility. Potassic to ultrapotassic melt
42 inclusions in UHT granulites suggests that lower crustal anatexis may play a stronger role in
43 the redistribution of heat-producing elements (such as K_2O), potentially influencing the thermal
44 structure of the continental crust.

45

46

47

48 1. INTRODUCTION

49 Buoyant andesitic continental crust (e.g. Rudnick, 1995; Taylor & McLennan, 1985) is a
50 distinctive feature that differentiates the Earth from all the terrestrial planets of the Solar
51 System. Although the upper levels of continents can be easily studied and sampled, the
52 structure and chemical composition of the lower continental crust remain mostly enigmatic,
53 due to its inaccessibility for direct observations. Rock xenoliths entrained in lavas and
54 tectonically exposed metamorphic terranes, characterized by mineral assemblages that record
55 lower crustal pressures (i.e. 5-15 kbar), are the only opportunities to obtain useful petrological
56 information on the deep crust. A large number of such exposed terrains has provided a strong
57 evidence that, on a regional scale, the lower continental crust can attain extreme thermal
58 regimes (> 775 °C/GPa; e.g. Harley, 2008; Kelsey and Hand 2015; Brown and Johnson, 2018).
59 Under these conditions, rocks are subjected to ultra-high temperature (UHT) metamorphism,
60 in which peak temperatures exceed 900 °C at typical pressures of 7-13 kbar (Harley, 1998),
61 with important implications for the geochemical differentiation, reworking and rheology of the
62 lower crust (Brown, Korhonen, & Siddoway, 2011; Jamieson, Unsworth, Harris, Rosenberg,
63 & Schulmann, 2011). UHT metamorphism is typically preserved in Mg-Al-rich metapelitic
64 granulites (Brandt, Will, & Klemd, 2007; Harley, Hensen, & Sheraton, 1990; Harley, 2008;
65 Kelsey, 2008; Santosh, Tsunogae, Li, & Liu, 2007), whose diagnostic paragenesis is
66 characterized by the presence of sapphirine + quartz, Al-rich orthopyroxene + sillimanite and
67 osumilite-bearing mineral assemblages (Harley, 1998; Kelsey, 2008). Progressing towards
68 UHT conditions, the lower crust buffers the increasing temperature undergoing extensive
69 partial melting, with the production of felsic magma that may then segregate from its source,
70 leaving behind a more mafic and less fertile residuum (Clemens & Stevens 2016; Sawyer 1994;
71 Sawyer, Cesare, & Brown, 2011; Vielzeuf & Holloway, 1988; White & Powell, 2002).
72 Although S-type granites and granitic leucosomes are the two most prominent products of

73 crustal anatexis, they do not provide the pristine chemical composition of the felsic anatectic
74 melt, because they are affected by fractional crystallization, crystal accumulation, chemical
75 contamination and presence of xenocrysts (Carvalho, Sawyer, & Janasi 2016; Marchildon &
76 Brown 2001; Milord, Sawyer, & Brown, 2001; Sawyer, 2008; Sawyer et al., 2011; Stevens,
77 Villaros, & Moyen, 2007). Consequently, initial anatectic melt compositions have been mainly
78 deduced from experimental studies (e.g. Clemens, Holloway, & White, 1986; Le Breton &
79 Thompson, 1988; Montel & Vielzeuf, 1997; Stevens, Clemens, & Droop, 1997) and
80 thermodynamic calculations (Grant, 2009; Holland & Powell, 2001; White, Stevens, &
81 Johnson, 2011). Another compelling method to determine the composition of crustal melts
82 during the early stages of anatexis is represented by primary melt inclusions entrapped in
83 peritectic minerals during incongruent melting of metapelitic rocks (Acosta-Vigil, Cesare,
84 London, & Morgan, 2007; Acosta-Vigil et al., 2016; Bartoli et al., 2013; Bartoli, Acosta-Vigil,
85 Tajčmanová, Cesare, & Bodnar, 2016; Cesare, Marchesi, Hermann, & Gómez-Pugnaire, 2003;
86 Cesare, Ferrero, Salvioli-Mariani, Pedron, & Cavallo, 2009; Ferrero et al., 2012; Ferrero,
87 Wunder, Walczak, O'Brien, & Ziemann, 2015). Primary melt inclusions (MI) in high-grade
88 metamorphic rocks typically show variable degrees of crystallization, ranging from glassy to
89 fully crystallized, the latter also referred as *nanogranitoids* (Cesare, Acosta-Vigil, Bartoli, &
90 Ferrero, 2015). Since MI represent droplets of early crustal melt, they preserve precious
91 information on its primordial composition, before the melt is subjected to any chemical change
92 due to magmatic differentiation.

93 At present, only very few studies have approached a detailed investigation of primary melt and
94 fluid inclusions at UHT conditions (e.g. Cesare et al., 2009; Ferrero et al. 2012; Tacchetto et
95 al., 2019) and therefore the pristine composition of anatectic melts and fluids in UHT rocks is
96 largely unconstrained. In order to overcome this paucity, we report the occurrence of primary
97 MI in peritectic minerals observed in UHT granulites of the Gruf Complex (Central Alps). The

98 aim of this study is to provide a microstructural framework and geochemical characterization
99 of anatectic melts and fluids and evaluate their role during crustal reworking processes
100 associated to UHT metamorphism.

101

102 2. GEOLOGICAL SETTING

103 The Penninic units of the Alpine nappe stack in the Central Alps are characterized by mineral
104 isograds, isotherms and isobars that have a concentric shape, defining the so-called “Lepontine
105 Metamorphic Dome” (Todd & Engi, 1997; Wenk, 1955). The Gruf Complex is a ~12 x 10 km
106 migmatitic body between Switzerland and northern Italy, that is located in the south-eastern
107 part of the Lepontine Dome (see Figure 1). To the west the Gruf Complex is delimited by the
108 Adula Nappe, whereas to the north is surrounded by the ultramafic bodies of the Chiavenna
109 Unit and by the crystalline rocks of the Tambo Nappe. To its eastern and southern margin, the
110 Gruf Complex is intruded by the Bergell pluton, a tonalitic to granodioritic calc-alkaline
111 intrusive body that formed between 33 and 30 Ma (Gianola et al., 2014; Oberli, Meier, Berger,
112 Rosenberg, & Gieré, 2004; Samperton et al., 2015; von Blanckenburg, 1992). The southern
113 extremity of the Gruf Complex, and part of the Bergell intrusion, are then cut by the 24 Ma old
114 Novate granite (Liati, Gebauer, & Fanning, 2000), a strong peraluminous S-type granitic stock
115 composed by a swarm of dykes that presumably derived from the partial melting of the
116 surrounding gneissic units (von Blanckenburg, Früh-Green, Diethelm, & Stille, 1992). The
117 dominant lithologies of the Gruf Complex are migmatitic orthogneisses, paragneisses and
118 micaschists (Galli et al. 2011), which underwent upper amphibolite facies migmatization
119 (Bucher-Nurminen & Droop 1983; Nagel, De Capitani, & Frey, 2002) between 34 and 29 Ma
120 (Galli et al. 2012). These rocks are also associated with irregular tabular bodies of charnockites
121 and leucogranites (Galli et al. 2011). Both migmatitic orthogneisses and charnockites are
122 characterized by the presence of rare sapphirine-bearing granulites, which mainly can be

123 observed as residual enclaves and schlieren (Galli et al. 2011). According to Galli et al. (2011),
124 granulites and charnockites formed at ultra-high temperature conditions of $T = 920\text{-}940\text{ }^{\circ}\text{C}$ and
125 $P = 8.5\text{-}9.5\text{ kbar}$, while migmatization of the surrounding gneissic and metapelitic lithologies
126 took place at $\sim 740\text{ }^{\circ}\text{C}$ and $6.5\text{-}7.5\text{ kbar}$ (Galli, Le Bayon, Schmidt, Burg, & Reusser, 2013).

127

128 **3. ANALYTICAL TECHNIQUES AND EXPERIMENTAL METHODS**

129

130 Back-scattered electron (BSE) images and semi-quantitative energy dispersive spectroscopy
131 (EDS) analyses of MI were carried out with a CamScan MX2500 Scanning Electron
132 Microscope (SEM), at the Department of Geosciences of the University of Padova (Italy).

133 Micro-Raman spectroscopy of MI was conducted with a HORIBA Jobin-Yvon LabRAM HR
134 800, equipped with a Nd:YAG laser ($\lambda = 532\text{ nm}$, laser power on sample $\sim 2\text{-}3\text{ mW}$), at the
135 Institute of Earth and Environmental Science, University of Potsdam. Raman spectra were
136 acquired between 100 and 4000 cm^{-1} with an acquisition time of 20 s during three spectral
137 accumulations. The typical setup also included a grating of 300 lines/mm , a slit width of 100
138 μm and a confocal hole set at 200 . Spectral resolution is considered to be of $\sim 10\text{ cm}^{-1}$, compared
139 to available literature and databases.

140 The chemical composition of garnets was measured with a CAMECA SX50 microprobe,
141 equipped with 4 wavelength dispersive spectrometers, at the C.N.R.-I.G.G. (Consiglio
142 Nazionale delle Ricerche – Istituto di Geoscienze e Georisorse), Department of Geosciences,
143 University of Padova. Analyses were carried out using a focussed beam with a current of 20
144 nA and an accelerating voltage of 15 kV . Counting times for each element were 10 s on peak
145 and 5 s for the background.

146 Major element composition of glass and minerals in the re-homogenized and glassy MI were
147 determined with a JEOL JXA 8200 microprobe at the Department of Earth Sciences

148 (University of Milano, Italy) employing an acceleration voltage of 15 kV, a beam current of 5
149 nA and beam diameter of 1 μm . Acquisition times were 10 s on peak and 5 s for background,
150 measuring Na and K first to avoid diffusional losses. Alkali concentrations were corrected
151 using conservative factors obtained by the analysis of hydrous and anhydrous leucogranitic
152 glasses of known composition. In particular, the standards employed in this study were a 10.1
153 wt% H₂O-bearing glass (LGB 2; Behrens & Jantos, 2001), a 5.5 wt% H₂O-bearing glass (DL;
154 Acosta-Vigil, London, Morgan, & Dewers, 2003) and a nearly anhydrous (H₂O = 300 \pm 42
155 ppm) glass (B; Morgan & London, 2005).

156 Single garnet core and rim fragments (~1.5 x 2 mm in size), used for the re-homogenization
157 experiments of the nanogranitoids, were manually separated by cutting from 1.5 mm-thick
158 double-polished thick sections. Two sets of experiments were performed. In the first set, garnet
159 chips were charged into Au capsules (external diameter = 3 mm) alternating them with
160 powdered graphite (granulometry <45 μm). In the second type of experiments, the same gold
161 capsules were filled with alternating layers of garnet chips and powdered SiO₂, obtained from
162 tetraethylorthosilicate (TEOS). In both cases, capsules were crimped and sealed with an arc-
163 welder. Experiments with garnet + graphite charges were conducted using single-stage and
164 end-loaded piston cylinders at the Laboratory for Experimental Petrology, Department of Earth
165 Sciences, University of Milano. Capsules were placed vertically into MgO spacers and then
166 inserted in a NaCl sleeve-pyrex-graphite piston cylinder assembly. The temperature was
167 measured with a K-type (nickel-chromium / nickel-alumel) thermocouple, with an estimated
168 accuracy of ± 5 °C and no corrections for the pressure effects on the electromotive force were
169 applied. Experiments were performed at 10 - 12 kbar (i.e. higher than the calculated peak
170 conditions, in order to prevent decrepitation of the inclusions) between 950 °C and 800 °C and
171 with variable run durations (1 to 10 hours). This allowed us to constrain the interval in which
172 re-homogenization was best attained. All runs were first pressurized to 10 - 12 kbar at room

173 temperature and then the assembly was isobarically heated at a constant heating rate (100
174 °C/min) until the targeted temperature was achieved. The experiments were terminated by
175 switching off the power supply, resulting in quenching rates of ~40 °C/s. Capsules were then
176 dissolved in aqua regia and the recovered garnets were singularly mounted in epoxy.
177 Experiments with garnets + quartz charges were performed at the Institute of Geochemistry
178 and Petrology, ETH Zurich, using single stage and end-loaded piston cylinders. The assembly
179 consisted in a talc sleeve, pyrex (only for end-loaded piston cylinder), a graphite furnace and
180 MgO spacers, in which the Au capsules were positioned. Temperature was measured with a B-
181 type (Pt₉₄Rh₆ – Pt₇₀Rh₃₀) thermocouple. Experiments were performed at 12 kbar, 875 °C and
182 with a duration of 10 h. The experimental procedure was the same used for the experiments
183 with garnets + graphite charges. Although the two set of experiments have slightly different
184 assemblies, it is assumed that this has no relevant influence on the re-homogenization of the
185 inclusions.

186 H₂O and CO₂ contents of remelted inclusions were determined by Nano Secondary Ion Mass
187 Spectrometry (NanoSIMS) employing a Cameca NanoSIMS 50 at Muséum National d'Histoire
188 Naturelle of Paris, following the approaches of Bartoli, Cesare, Remusat, Acosta-Vigil, and
189 Poli (2014) and Créon, Levresse, Remusat, Bureau, and Carrasco-Núñez (2018). Polished
190 experimental capsules with quartz-garnet charges and standard glasses were mounted in
191 indium, to lower the hydrogen background in the analysis chamber (Aubaud et al., 2007), and
192 coated with gold (20 nm thick). Before each analysis, a 5 x 5 µm surface area was initially pre-
193 sputtered for 150 s with a 170 pA primary Cs⁺ rastering beam, in order to remove the gold
194 coating and reach a sputtering steady-state (Thomen, Robert, & Remusat, 2014). Subsequently,
195 the primary Cs⁺ beam was set at 17 pA and scanned over a surface area of 3 x 3 µm.
196 Nevertheless, to avoid surface contamination, only ions from the inner 1 x 1 µm region were
197 collected with the “beam blanking mode”. Exposed MI were analysed through collection of

218 secondary ions of $^{12}\text{C}^-$, $^{16}\text{OH}^-$, $^{28}\text{Si}^-$ and $^{56}\text{Fe}^{16}\text{O}^-$ in multicollection mode. Mass resolving power
219 was set at minimum 5500, enough to resolve interferences on $^{16}\text{OH}^-$. Each analysis consisted
220 in a stack of 200 cycles, with a duration of 1.024 s for every single cycle. The vacuum in the
221 analysis chamber never exceeded 2.5×10^{-10} Torr during the entire session of measurements.
222 H_2O and CO_2 contents of the glasses were quantified measuring $^{16}\text{OH}^-/^{28}\text{Si}^-$ and $^{12}\text{C}^-/^{28}\text{Si}^-$ ratios,
223 respectively (Bartoli et al., 2014, Créon et al., 2018). These ratios were converted into
224 concentrations using calibration curves obtained from standards of known composition.
225 Standards for the determination of H_2O included two standards used for the alkali correction
226 (DL and B, see above) plus an additional hydrous glass with 4.3 wt% H_2O (LGB1; Behrens &
227 Jantos, 2001). On the other hand, standards for CO_2 consisted in four trachyandesites (STR 9,
228 10, 11 and 13) from the Stromboli volcano, experimentally doped in carbon and water by
229 Bureau et al. (2003). Concentrations and uncertainties were calculated using the R program,
230 following the procedure described in Bartoli et al. (2014) and Thomen, Robert, & Remusat
231 (2014). Average major element and volatile concentrations of the analysed MI are reported in
232 Table 1.

233

234 4. RESULTS

235

236 4.1 Mineral assemblages of the UHT granulites

237 All the investigated samples were collected in the lower part of Val Codera, near the locality
238 of Bresciadega (see also Galli et al., 2011). Two varieties of UHT granulite with granofelsic
239 texture were studied (Figure 2). The first type corresponds to type A of Galli et al. (2011) and
240 consists in a dark, massive granulite containing large garnet (up to 2 cm) and sapphirine (up to
241 5 mm) porphyroblasts, associated with a matrix of up to 3 mm-sized orthopyroxene (Opx),
242 cordierite (Crd), biotite (Bt) and K-feldspar (Kfs). Garnet (Grt) appears as faint pink, rounded

223 crystals showing a clear zonation. Numerous mineral inclusions of biotite, orthopyroxene,
224 sapphirine, plagioclase (Pl), cordierite, rutile (Rt), ilmenite, sillimanite, tourmaline, apatite
225 (Ap), spinel, staurolite, corundum, monazite and zircon can be observed within the
226 porphyroblastic garnets. On the other hand, sapphirine (Spr) is generally hypidiomorphic,
227 prismatic and pleochroic (from pale-red to blue), showing sometimes a longitudinal, simple
228 twinning system.

229 The second type of granulite belongs to type C of Galli et al. (2011). The rock is massive and
230 melanocratic with a mineral assemblage made of porphyroblastic garnet (up to 1.5 cm),
231 porphyroblastic orthopyroxene (<1.5 mm), biotite, cordierite, sillimanite and symplectitic
232 spinel. Sapphirine may be present as symplectitic grains in the rock matrix, or as small (<500
233 μm) inclusions within garnet. Other mineral inclusions observed within garnet grains are:
234 biotite, quartz, sillimanite, cordierite, plagioclase, spinel, zircon, apatite, corundum, rutile,
235 staurolite and tourmaline.

236 In both granulites, melt inclusions (MI) are mainly present in the porphyroblastic garnets,
237 where they may occur as single, scattered inclusions or may form mm-large clusters. These
238 clusters are commonly found in the cores of the garnets (Figure 3a-b), but their occurrence
239 towards to the rims has been also noticed (Figure 3c-d). The presence of different birefringent
240 crystals under crossed polars (Figure 3e-f) indicates that the majority of MI are polycrystalline
241 (i.e. nanogranitoids; Cesare et al. 2015). Often, fluid inclusions, usually showing a distinctive
242 darker appearance and high birefringence (Figure 3g), occur associated to nanogranitoids
243 within the same clusters. Moreover, sporadic glassy inclusions containing shrinkage bubbles
244 have been also observed (Figure 3h). In contrast to polycrystalline MI, glassy MI occur
245 exclusively as single inclusions dispersed in the garnets, without forming clusters, although
246 occasionally, glassy MI may occur close to nanogranitoids. Moreover, rare MI with multiple
247 birefringent phases have been also identified in sapphirine porphyroblasts of Type A granulites

248 (Figure 3i). The MI in sapphirine are too rare to allow their experimental re-melting, but were
249 characterized by Raman spectroscopy.

250

251 **4.2 Micro-Raman characterization of glassy, nanogranitoid and fluid inclusions**

252 Micro-Raman investigations for the glassy inclusions observed in Type A samples show that
253 the glass is hydrous (see Figure 4a). Furthermore, the single Raman band displayed at 1382
254 cm^{-1} (Figure 4a) indicates that some dissolved molecular CO_2 is also present in the melt
255 (Brooker, Kohn, Holloway, McMillan & Carroll, 1999; Ni & Keppler 2013). Shrinkage
256 bubbles in the glass contain CO_2 , as demonstrated by the presence of the “Fermi-doublet” in
257 the Raman spectra (see Figure 4a). Densities of CO_2 in these bubbles (see Table S1), calculated
258 with the densimeter of Wang et al. (2011), range between 0.5 and 0.8 g/cm^3 ($\pm 0.3 \text{ g/cm}^3$).
259 Raman analyses showed that nanogranitoids (Figure 4b,c) may contain quartz, muscovite,
260 phlogopite, K-feldspar, kokchetavite (a polymorph of KAlSi_3O_8) and an unknown phase with
261 a main Raman peak at 430 cm^{-1} . Unexpectedly, Raman investigations also showed that some
262 of these nanogranitoids could retain some CO_2 bubbles (Figure 4b,c), suggesting the existence
263 of micropores. Nanogranitoids observed in sapphirine crystals from type A granulites (Figure
264 4d) contain quartz, muscovite, kokchetavite and the unknown phase with the peak at 430 cm^{-1} .
265 Fluid inclusions (FI), present in both granulites, display a fluid fraction as well as a solid
266 portion, in which several phases were identified. In type A granulites (together with CO_2)
267 muscovite, Fe-magnesite, calcite, pyrophyllite, quartz and biotite were the most common
268 phases. On the other hand, type C granulites have FI composed of – in addition to the same
269 phases observed for type A – paragonite, magnesite, siderite, phlogopite, N_2 and CO (Figure
270 4e,f). For one inclusion it was possible to calculate the relative amount of components in the
271 fluid, applying the method of Dubessy, Poty, and Ramboz (1989). The obtained molar fractions
272 were 94 mol% for CO_2 and 6 mol% for N_2 . For what concerns the carbonate phases, Fe-

273 magnesite in FI of both granulites has variable Fe content ($Fe\# = 100 \times Fe/(Fe + Mg + Mn +$
274 $Ca + Al)$, as defined by Boulard, Guyot, & Fiquet, 2012), typically ranging between $Fe\# = 20$
275 and $Fe\# = 55$. Calculated CO_2 densities (Wang et al. 2011) for FI in type A granulites vary
276 between 0.3 and 1.0 g/cm^3 (see Table S1), a range comparable to that calculated for type C
277 granulites (0.4-0.8 g/cm^3). It is also remarkable to notice that, neither in the shrinkage bubbles
278 of glassy inclusions nor in fluid inclusions, H_2O was detected.

279

280 4.3 Microstructures of nanogranitoids

281 Detailed observations with SEM allowed to determine the principal microstructural features of
282 nanogranitoids, as well as their mineral assemblage. Typically, MI of both Type A and C
283 granulites have an elliptical faceted shape, with major axes ranging between 2 and 115 μm
284 (Figure 5a-f). Nevertheless, for Type A, the size of MI decreases from rims to cores. MI in the
285 rims have an average size of $47 \pm 5 \mu m$, those found in the intermediate region of the garnets
286 (i.e. between cores and rims) are $35 \pm 5 \mu m$, while MI found in the cores have an average size
287 of $24 \pm 3 \mu m$. Together with nanogranitoids with a nearly negative crystal shape (Figure 5a,b),
288 some MI display natural decrepitation, with offshoots that are arranged radially around the
289 inclusions (Figure 5c,d). The number of offshoots per inclusion varies drastically (from 1 to
290 10), with larger inclusions showing more (and usually longer) offshoots. The calculated
291 average length of the offshoots for MI $<45 \mu m$ in size is 6 μm , whereas MI bigger than 45 μm
292 have an average length of the offshoots of 13 μm . Glassy inclusions in Type A granulites show
293 negative crystal shapes and their size is $<15 \mu m$ (average diameter: 8 μm). MI in type C
294 granulites also have an elliptical faceted shape and show a polycrystalline nature. However, in
295 contrast to MI in type A granulites, they have similar sizes from core to rim and hardly exceed
296 40 μm in diameter, with an average length of the major axis of $21 \pm 2 \mu m$. Also garnets from

297 type C granulites show some decrepitated MI (Figure 5e,f), which have an average length of
298 the offshoots of 5 μm .

299 In general, the mineral assemblage within nanogranitoids has an inequigranular and interlobate
300 fabric composed of mainly hypidiomorphic crystals of quartz, biotite, muscovite, plagioclase,
301 K-feldspar and very rarely aluminosilicates (Figure 5a-f). Sometimes nanogranitoids clearly
302 display an igneous microstructure defined by intergrowths of quartz, feldspars and biotite. SEM
303 analyses also highlighted the presence of accessory minerals, typically Cl-bearing apatite and
304 rutile, which are usually more idiomorphic compared to the other phases (Figure 5d,e).

305

306 **4.4 Chemical composition of host garnets and minerals in nanogranitoids**

307 In this section we report only chemical analyses of garnet porphyroblasts. For detailed chemical
308 analyses of all rock-forming minerals see Galli et al. (2011). Analysed garnets of Type A and
309 Type C granulites are almost pure almandine-pyrope solid solutions and display a zonation
310 marked by pyrope-richer rims (Type A = $\text{Alm}_{43-50}\text{Prp}_{44-53}\text{Sps}_{0-2}\text{Grs}_{2-6}$; Type C = $\text{Alm}_{44-54}\text{Prp}_{43-}$
311 $_{55}\text{Sps}_{0-1}\text{Grs}_{1-3}$) compared to cores (Type A = $\text{Alm}_{47-52}\text{Prp}_{39-48}\text{Sps}_{1-3}\text{Grs}_{3-9}$; Type C = Alm_{50-}
312 $_{62}\text{Prp}_{32-47}\text{Sps}_{0-1}\text{Grs}_{2-6}$).

313 For what concerns the nanogranitoids, their size often hampers a precise chemical
314 characterization of their mineral assemblage. Nevertheless, some nanogranitoid inclusions in
315 Type A granulites were large enough to allow the chemical analysis of the single mineral
316 phases (see Table S2). Plagioclases show variable X_{An} [$\text{An} / (\text{An} + \text{Ab})$] between 0.10 (albite)
317 and 0.49 (andesine). By contrast, biotites are Ti-free and characterized by X_{Mg} [$\text{Mg} / (\text{Mg} +$
318 $\text{Fe}^{2+})$] ranging from 0.71 to 0.86. Other K-rich phases that have been recognized are K-feldspar
319 and phengite (Ph), the latter displaying a $X_{\text{Mg}} = 0.59$.

320

321

4.5 Remelted nanogranitoids

322 Experiments between 900 and 875 °C produced the most homogenized glasses. Remelted MI
323 are typically <15 µm in size and display elongated to perfect negative crystal shapes (Figure
324 6a-d). Some inclusions, together with glass, display mineral phases like rutile and apatite that
325 indent the walls of the MI (Figure 6c). These phases are interpreted to be accidental minerals
326 trapped during the formation of the inclusions, since they are observed before and after the re-
327 homogenization experiments. Therefore, they are not daughter minerals (i.e. phases that
328 crystallized directly from the melt; e.g. Ferrero et al., 2012; Acosta-Vigil et al. 2016).
329 Moreover, few MI may also exhibit bubbles (Figure 6d), suggesting that the fluid underwent
330 incomplete dissolution in the melt during the experimental run or alternatively it diffused
331 during cooling to form shrinkage bubbles (Lowenstern, 1995). A little fraction of the analysed
332 MI population is characterized by the presence of unmelted daughter minerals, testifying
333 incomplete remelting (Figure 6e,f). Typical daughter minerals coexisting with glass are
334 euhedral to subhedral biotite grains ($X_{Mg} = 0.82 - 0.88$ and $TiO_2 < 0.2$ wt%, see Table S3) and
335 rounded quartz crystals.

337 Experiments with garnets + quartz charges at 875 °C produced substantial amounts of melt in
338 the matrix outside the garnets (Figure 6g), that infiltrated within the garnet crystals and led to
339 the formation of large pockets of melt. These melt pockets differ microstructurally from
340 remelted MI by their size (commonly >20 µm in diameter, although few of them have sizes
341 comparable to remelted MI) and by the striking abundance of bubbles, a feature that indicates
342 high fluid contents (Figure 6h). The matrix melt derives from the melting of mineral
343 intergrowths or matrix phases, such as biotite and feldspar, which could not be avoided during
344 the manual crushing of the garnet chips. Often, a peritectic reaction between the melt and the
345 garnet can be observed, with the formation of orthopyroxene or corundum (Figure 6g).

346 Evidence of overheating in the MI has been observed at all temperatures, but predominantly at
347 950 °C. This is indicated by the formation of peritectic phases such as orthopyroxene or gedrite
348 (Figure 6i), by the recrystallization of new garnet within the inclusion or by a change in the
349 chemical composition of the garnet around the remelted inclusions. Chemical analyses (see
350 Table S3) for peritectic orthopyroxenes indicate that they are Al-rich (8.3 – 10.4 wt% at 950
351 °C and 6.5 – 9.2 wt% Al₂O₃ at lower temperature), with X_{Mg} = 0.69 - 0.73 (at 950 °C) and 0.67
352 – 0.84 (at 900-875 °C). These compositions are comparable to those measured in the
353 orthopyroxene porphyroblasts in the matrix of granulites (Galli et al. 2011). One analysis of a
354 recrystallized garnet 950 °C shows that the new garnet is Fe-poorer and Mg-richer (X_{Mg} = 0.64)
355 compared to the original garnet composition (X_{Mg} = 0.47). Since at 950 °C microstructural
356 evidence for overheating is dominant, the glasses analysed at these conditions were considered
357 not representative and therefore disregarded (Figure S1). For what concerns the experiments
358 performed at 800 °C, MI often display incomplete remelting and only rare homogeneous
359 glasses could be found. This might indicate that this temperature corresponds to the lower
360 boundary for remelting experiments.

361

362 4.6 Chemical composition of glasses

363 Figure 7a-b and Table S4 report chemical analyses of glasses for remelted inclusions and for
364 preserved glassy inclusions, all recalculated on anhydrous basis. For what concerns
365 experimentally remelted MI, analyses that displayed clear evidence for
366 contamination/interaction with the host garnet, or mixing with matrix melt, were discarded and
367 were not discussed (see Figure S1). Glasses are dominantly peraluminous (ASI = 1.0 – 1.6,
368 [Al₂O₃/(CaO+Na₂O+K₂O)]_{molar}), with only two MI that fall into the peralkaline field. Natural
369 glassy inclusions have a slightly more restricted range (ASI = 1.0 – 1.1) compared to re-
370 homogenized nanogranitoids. With rare exceptions, analysed MI are corundum-normative (Crn

371 = 0.2 – 4.88, see Table S4). Both re-homogenized MI and glassy inclusions are predominantly
372 rhyolitic in composition, with $\text{SiO}_2 = 68.6 - 79.7 \text{ wt\%}$ and $\text{Na}_2\text{O}+\text{K}_2\text{O} = 5.3 - 12.0 \text{ wt\%}$ (on
373 anhydrous basis), developing a negative correlated trend in the SiO_2 vs. $\text{Na}_2\text{O}+\text{K}_2\text{O}$ space
374 (Figure 7b). However, melts with a trachytic composition have been also observed, as shown
375 by one MI from Type A granulite remelted at $875 \text{ }^\circ\text{C}$. Glassy inclusions have slightly higher
376 total alkali contents compared to remelted nanogranitoids. Plotted in Harker variation diagrams
377 (Figure 8 a-f), glassy and re-homogenized inclusions show a clear increase in Al_2O_3 and K_2O
378 with decreasing SiO_2 . By contrast, CaO , $\text{FeO}+\text{MgO}+\text{TiO}_2$, Na_2O and Cl display more scattered
379 distributions. Glassy inclusions differ from re-homogenized nanogranitoids by their low CaO
380 ($<0.2 \text{ wt\%}$ vs. $0.2\text{-}2.0 \text{ wt\%}$) and $\text{FeO}+\text{MgO}+\text{TiO}_2$ ($<1.4 \text{ wt\%}$ vs. $1.4\text{-}3.4 \text{ wt\%}$) concentrations.
381 All the analysed melt inclusions are characterized by a decoupling in the alkali content, with
382 most inclusions showing Na_2O concentrations $<2 \text{ wt\%}$ and $\text{K}_2\text{O} > 6 \text{ wt\%}$. Re-homogenized MI
383 have variable K\# ratios [$\text{K\#} = \text{molar K}_2\text{O}/(\text{K}_2\text{O}+\text{Na}_2\text{O}) \times 100$], ranging from 72 to 98 (average:
384 87 ± 4) for Type C granulites and from 56 to 82 (average: 71 ± 8) for Type A granulites.
385 Similarly, glassy inclusions from Type A granulites have K\# between 59 and 87 (average: 77
386 ± 5).

387 H_2O and CO_2 concentrations of remelted inclusions, determined by NanoSIMS, are reported
388 in Table 2 and shown in Figure 9. H_2O contents in the analysed MI range from 2.9 to 8.8 wt%,
389 without any clear distinction between garnet cores and rims. Indeed, calculated averages are
390 $6.2 \pm 1.3 \text{ wt\%}$ (type A) and $7.1 \pm 3.0 \text{ wt\%}$ (type C) for garnet cores, whereas for garnet rims is
391 4.4 ± 0.9 (type C). By contrast, CO_2 concentrations allow to subdivide the analysed MI into
392 two distinct families, one clustering at $\sim 1200 \text{ ppm}$ and the other showing concentrations <500
393 ppm. From Figure 9 it is also possible to notice that MI with low CO_2 concentrations are
394 characterized by a narrower range in H_2O ($4.1 - 5.5 \text{ wt\%}$) compared to MI with higher CO_2
395 contents.

396 5 DISCUSSION

397 5.1 Anatexis at UHT conditions

398 The mineral assemblage displayed by nanogranitoid inclusions within the peritectic garnet of
399 the studied granulites indicates that these inclusions represent droplets of pristine felsic melt,
400 an observation that was also confirmed by the measurement of their chemical composition after
401 remelting experiments. Therefore, the investigated MI allow firstly an appraisal of the melting
402 regime on the regional scale and secondly, a better constraint on crustal anatexis under UHT
403 conditions.

404 Granulites from the Gruf Complex are characterized by the presence of glassy and
405 nanogranitoid inclusions. Their coexistence at the micrometric scale indicates that the melt
406 behaves differently when it is trapped within pores. Glassy inclusions are on average smaller
407 than nanogranitoids and therefore their preservation may be explained by inhibition of
408 crystallization (Cesare et al., 2009) due to the greater supersaturation required by smaller pores
409 to develop crystals compared to larger ones (Holness & Sawyer, 2008). Alternatively, lower
410 H₂O contents and, in turn, higher viscosities in some inclusions could have inhibited the
411 nucleation with the formation of glass (Bartoli, Acosta-Vigil, & Cesare, 2015). A different
412 volatile content in glassy and crystallized melt inclusions seems to be supported by the different
413 electron microprobe totals (Table 1). The chemical composition of re-homogenized MI is
414 consistent with the mineral assemblage observed in the nanogranitoids. This can be also
415 observed in the AKF diagram (Figure 10), where analysed glasses for Type A granulites plot
416 within the triangle defined by the tie-lines of the minerals measured in the non-remelted
417 nanogranitoids. Although the chemical composition of the minerals forming MI in Type C
418 granulites could not be measured (Figure 10b), it is assumed that the mineral assemblage should
419 be similar to the one noticed for Type A granulites and therefore an equivalent triangle for MI
420 in Type C granulites is expected. From Figure 10a it is also possible to observe that glassy

421 inclusions plot towards the K-apex of the triangle, while remelted MI tend to be more scattered,
422 with some analyses plotting slightly outside the triangle defined by the mineral assemblage. In
423 the Qz-Ab-Or normative diagram (see Figure S2) glassy and re-homogenized inclusions from
424 the Gruf Complex plot close to the Qz-Or join, considerably away from the haplogranite ternary
425 eutectic. Furthermore, MI also plot at lower Ab-contents compared to charnockites,
426 leucogranites and S- and A-type granites, but overlap the field displayed by MI from the Kerala
427 Khondalite Belt, consistent with their UHT nature.

428 Figure 8 shows that the analysed glasses have markedly different compositions compared to
429 the charnockites of the Gruf Complex, indicating that the charnockites might have been
430 generated from another metapelitic source or during a melting event unrelated with the
431 formation of the granulites. By contrast, MI have similar concentrations for CaO, Al₂O₃ and
432 FeO+MgO+TiO₂ compared to the leucogranites that intrude the migmatitic paragneisses and
433 micaschists (see Galli, Le Bayon, Schmidt, Burg, & Reusser, 2013). Moreover, MI overlap the
434 trend displayed by the melt inclusions from the Kerala Khondalite Belt (Cesare et al. 2009;
435 Ferrero et al. 2012), even though the latter have slightly higher FeO+MgO+TiO₂ and Cl
436 concentrations. On the other hand, MI from the Gruf Complex have different trends for
437 FeO+MgO+TiO₂, alkalis, and partly CaO, compared to A- and S-type granites. This evidence
438 strongly reinforces the assumption that S- and A-type granites do not mirror primary crustal
439 melts, but liquids modified by a large plethora of mechanisms during their production and
440 segregation from the source (see Introduction).

441 MI are characterized by high K₂O and low Na₂O concentrations compared to regional
442 charnockites and leucogranites, but also relative to A- and S-type granites (Figure 8d-e).
443 However, a decoupled behaviour in alkali contents has been also noticed in the MI from the
444 Kerala Khondalite Belt (KKB, S-India), in which ultrapotassic rhyolitic melts have been
445 reported (Cesare et al. 2009; Ferrero et al., 2012). It is generally believed that khondalites from

446 the KKB originated during crustal anatexis at UHT conditions (Nandakumar and Harley, 2000;
447 Yu, Santosh, Li, & Shaji, 2019) and therefore it seems that high K_2O/Na_2O ratios are a
448 distinctive feature of anatectic melts at extremely high crustal temperatures. K-rich melts have
449 also been experimentally derived by Patiño Douce and Johnston (1991) from a pelitic starting
450 material. In particular, their experiments between $P = 7-13$ kbar and $T = 875-1075$ °C give
451 K_2O -rich and Na_2O -poor liquids that closely resemble MI from the Gruf Complex and the
452 KKB. Additionally, Droop, Clemens, & Dalrymple (2003) obtained K-rich melts (K_2O/Na_2O
453 $= 4.1 - 9.5$) from partial melting experiments of metapelitic micaschists at 900 °C and 5 kbar.
454 In the AKF diagram (Figure 10) analyses for the KKB, as well as the experimental glasses of
455 Droop et al. (2003) and Patiño-Douce & Johnston (1991), plot towards the K-apex, overlapping
456 in part the field formed by the MI of the Gruf Complex. The fact that K-rich melts are observed
457 up to UHT conditions, independently of the pressure, may be explained by the role of titanium
458 in expanding the stability field of biotite towards higher temperatures. Consequently, the
459 melting of biotite may be delayed, compared to the melting of sodic plagioclase, leading to an
460 enrichment of potassium in the melt.
461 All this evidence suggests that melts produced at the onset of anatexis under high to ultra-high
462 temperature conditions are dominantly potassic to ultrapotassic, as was also recognized in
463 previous studies (Cesare et al., 2009; Ferrero et al., 2012).

464

465 **5.2 Volatiles in the deep crust**

466 Primary clusters of melt and fluid inclusions occur typically in both garnet cores and rims of
467 the granulites from the Gruf Complex, indicating that they were entrapped during the same
468 anatectic event. The coexistence of CO_2 with stepdaughter phases such as carbonates (Fe-
469 magnesite, siderite and calcite) and phyllosilicates (pyrophyllite, muscovite and biotite) within
470 FI strongly suggests that the original fluid had a COH(N) composition. Nevertheless, the

471 presence of K-rich phases such as muscovite and biotite might also indicate that some of the
472 FI are instead mixed inclusions (fluid + melt) and therefore part of the H₂O may derive from
473 the melt. Similar mixed inclusions were documented in garnet of crustal enclaves (Cesare,
474 Maineri, Baron Toaldo, Pedron, & Acosta-Vigil, 2007) and anatectic granitoids (Ferrero,
475 Braga, Berkesi, Cesare, & Laridhi Ouazaa, 2014). Although H₂O as a free phase has been never
476 demonstrated by Raman spectroscopy within the measured FI of the Gruf Complex, its
477 occurrence on the nanometric scale cannot be excluded (see also Lamadrid, Lamb, Santosh, &
478 Bodnar, 2014; Esposito et al., 2016). On the other hand, carbonates and phyllosilicates are
479 interpreted as re-speciation products after post-entrapment carbonation and hydration reactions
480 between garnet host and fluid, as it has been suggested also for other HT/UHT terranes (Ferrero
481 et al. 2014; Tacchetto et al., 2019; Carvalho et al., 2019). Hence, the different densities
482 displayed by the FI are a direct consequence of incomplete reactions, which leave a variable
483 amount of CO₂ in the residual fluid.

484 Raman spectroscopy measurements allowed to confirm the presence of CO₂ and H₂O in glassy
485 and nanogranitoid inclusions (see Fig. 4), indicating that both inclusions derived from the
486 similar melt. The ubiquity of pure CO₂-rich shrinkage bubbles (without any apparent detectable
487 H₂O, see discussion above) within glassy inclusions can be explained by the preferential
488 partition of H₂O into silicate melt than CO₂ (e.g. Holtz, Johannes, Tamic, & Behrens, 2001;
489 Tamic, Behrens, & Holtz, 2001). Further investigations with NanoSIMS enabled the
490 quantification of the minimum volatile component dissolved in this melt. The dissolved H₂O
491 measured in the MI displays a wide spectrum (2.9-8.8 wt%), with a large proportions of the
492 analyses that have >5 wt%. Such values indicate that crustal melts at T ~ 900 °C can have
493 higher H₂O contents than commonly assumed for the haplogranitic system. However, as
494 pointed out by Holtz et al. (2001), this behaviour is to be expected for melts that markedly
495 differ from eutectic compositions and that involve the breakdown of micas in the melting

496 reaction, conditions which are both realised for the MI from the Gruf Complex. Moreover,
497 analogous H₂O contents have been also observed in MI from the granulites and upper
498 amphibolite-facies migmatites from the Strona Valley, in the Ivrea Zone (Carvalho et al.,
499 2019). It is therefore suggested that, at HT/UHT conditions, anatectic rhyolitic melts have
500 considerable but uneven H₂O contents, which may strongly affect the mobility of these
501 magmas. Variable water contents in anatectic melts were also recognized by Bartoli et al.
502 (2014) in peritectic garnets of amphibolite-facies migmatites close to the Ronda peridotite and
503 were interpreted to represent micro-scale buffered compositions due to heterogeneous domains
504 in the protholith. A similar mechanism might be envisaged also for the MI of the Gruf Complex,
505 since low and high values sometimes can be found in the same garnet crystal. Another striking
506 feature of the analysed MI is the marked difference in the CO₂ contents (Figure 9), with
507 inclusions forming two distinct groups. The highest CO₂ concentrations are nearly identical to
508 those observed in MI from granulites and amphibolites from the Ivrea Zone (see also Carvalho
509 et al., 2019), whereas the “CO₂-depleted” MI plot at lower values.

510 Our results clearly demonstrate that the peritectic garnets of the investigated UHT granulites
511 grew with coexisting COH-bearing fluids and silicate melts, that, due to the low solubility of
512 CO₂ in silicate melts (Holtz et al., 2001; Tamic et al., 2001), led to fluid-melt immiscibility.
513 The origin of the COH fluid in high-grade metamorphic rocks has been often source of debates.
514 The granulites from the Gruf Complex lack graphite in their mineral assemblage and therefore
515 an internal origin for carbon seems to be unlikely. However, the absence of graphite in the
516 rocks does not preclude the possibility that graphite was present in the original mineral
517 assemblage and was completely consumed before or during anatexis. Another source for
518 carbon may be represented by carbonaceous sediments interlayered in metapelitic rocks. In the
519 Gruf Complex very thin carbonate layers have been recognized only at the contact with the
520 ultramafic Chiavenna Unit, but never within the migmatitic gneisses that host the UHT

521 granulites and thus a sedimentary source for carbon does not seem a convincing argument.
522 Accordingly, we infer that CO₂ was externally introduced in the lower crust, very likely from
523 a mantle source. This is in agreement with previous studies that suggested the mantle as a
524 common source for CO₂ in high temperature granulites (e.g. Hoefs & Touret, 1975; Jackson,
525 Matthey, & Harris, 1988; Santosh & Omori, 2008; Touret & Huizenga, 2011). However, we
526 also emphasize that without a rigorous isotopic determination for carbon, the origin of CO₂
527 remains uncertain. On the other hand, the minor N₂ amount detected in Type C granulites is
528 interpreted to derive from the breakdown of NH₄⁺-bearing micas during anatexis (e.g. Moin,
529 Guillot, & Gibert, 1994; Cesare et al., 2007).
530 For the Gruf Complex, if an external source for CO₂ is assumed, the different CO₂
531 concentrations in the MI may be related to small-scale heterogeneities of the fluid composition
532 within the matrix surrounding the growing peritectic garnets.

533

534 **5.3 Rheology of anatectic melt**

535 Granulites from the Gruf Complex represent UHT residues of metapelitic lower crustal rocks,
536 attesting that substantial partial melting (~40 - 50%, Galli et al., 2011) occurred in the
537 lowermost portions of the continental crust. This partial melting generated rhyolitic melts that
538 are now preserved in the MI found in the peritectic garnets of the investigated granulites. An
539 important aspect related to the genesis of anatectic rhyolitic melts is to determine some
540 fundamental physical properties that govern the mobilization of these melts through the crust,
541 such as viscosity and density. The complete analysis (geochemistry and water content) of four
542 MI allowed us to calculate the viscosity of the melt. Assuming a temperature of 875 °C, which
543 corresponds to the experimental temperature, and applying the equation of Scaillet, Holtz,
544 Pichavant, & Schmidt (1996), viscosities vary between 10^{3.9} and 10^{4.5} Pa·s. These values are
545 comparable to those calculated using the equation of Giordano, Russell, & Dingwell (2008),

546 which range between $10^{4.1}$ and $10^{4.8}$ Pa·s and are ~2 orders of magnitude lower than for
547 commonly assumed nearly anhydrous crustal melts at 900 °C. The calculated viscosities for
548 the melt during incipient UHT anatexis are consistent (within error) with the experimental
549 values obtained by [Ardia, Giordano, and Schmidt \(2008\)](#) for rhyolitic melts between 5 and 15
550 kbar at similar water contents ([see Figure S3](#)). Moreover, viscosities for MI from the Gruf
551 Complex overlap the range of viscosities obtained by [Bartoli et al. \(2013\)](#) for metatexites within
552 the metasedimentary sequence at the contact with the Ronda peridotite, in the Spanish Betic
553 Cordillera.

554 Combining viscosities with the respective densities allowed us to speculate on the ascent
555 velocity of the melt from its source. Our calculations were based on a simple porous flow model
556 ([Turcotte & Schubert, 2002](#)) and assuming an average density of 3.1 g/cm^3 for the lower crust
557 ([Hacker, Kelemen, & Behn, 2015](#)), a porosity of 8% (which corresponds to the liquid
558 percolation threshold of [Vigneresse, Barbey, & Cuney, 1996](#)) and a grain size for the matrix of
559 1-2 mm. For an average melt viscosity of $10^{4.4 \pm 0.3}$ Pa·s and an average density of 2.3 g/cm^3
560 (determined with the program DensityX by [Iacovino and Till, 2019](#)), melt velocities of 5-83
561 mm/year were calculated ([see Table S5](#)). Hence, if only porous flow is considered, at least tens
562 of thousands of years are needed before the melt will move at distances >1 km. At high melt
563 fractions it is expected that the pervasively distributed melt might be channelized by porosity
564 waves (e.g. [Connolly & Podladchikov, 2015](#)), resulting in an enhanced melt migration rate.

565 Furthermore, it has to be stressed that the calculated velocities are relative to a static solid
566 matrix. However, melting in the lower crust occurs in a dynamic environment, in which
567 deviatoric stresses may largely increase the ascent velocity of magmas ([Sawyer 1994](#)).

568 The Gruf Complex is as an example of upward extruded lower continental crust ([Galli et al.](#)
569 [2012; Galli et al. 2013](#)), that most probably was exhumed as a crustal diapir and for which an
570 exhumation rate of ~2 mm/year was estimated ([Droop and Bucher-Nurminen, 1984](#)). This

571 velocity is lower compared to those calculated for the anatectic melt and therefore it might be
572 plausible that the melt (or at least part of it) may have been effectively segregated from the
573 ascending migmatitic body. However, it is also expected that melts with physical properties
574 similar to those of the Gruf Complex may undergo less segregation if they are produced in
575 faster exhumed crustal sections. Field observations and numerical modelling (e.g. Ganguly,
576 Dasgupta, Cheng, & Neogi, 2000; Gerya, Perchuk, & Burg, 2008; Whitney, Teyssier, & Fayon,
577 2004) indicate that hot and partially molten crust may rise with velocities up to 50 mm/year,
578 i.e. in the range of those calculated in this study. Therefore, on the small scale, melts may
579 segregate from the source (forming leucosomes), but on the large scale they would probably
580 move coupled with the ascending hot crust (Teyssier & Whitney, 2002). Consequently,
581 segregation processes in migmatitic crustal domains should be evaluated also taking into
582 account the exhumation rate of the crust, and not only the ascent velocity of the melt. Although
583 the proposed approach might be simplistic, it gives a first-order approximation on the
584 ascending behaviour of pristine anatectic melts at the onset of crustal anatexis under UHT
585 regime.

586

587 **6. CONCLUSIONS**

588 Evidence that the Earth's continental crust has the ability to attain, on a regional scale, ultra-
589 high temperatures has been widely reported. Under these conditions, middle and lower crust
590 undergo substantial modifications that have important consequences on their rheological
591 properties, which, in turn, may largely affect the mass transfer at the crust-mantle boundary.
592 Peraluminous, K-rich rhyolitic melt inclusions observed in garnet porphyroblasts of UHT
593 granulites from the Gruf Complex are the result of crustal anatexis, the latter driven by the
594 breakdown of biotite. The presence of primary melt and fluid inclusions with both a COH
595 volatile component suggests that anatexis initially started due to dehydration melting reactions

596 and was promoted by the influx of an external CO₂ + H₂O fluid. Although the source of CO₂
597 cannot be undoubtedly ascertained, we infer that a mantle origin for the carbon is a more likely
598 explanation. On the other hand, the high H₂O content measured in the melt inclusions suggests
599 that anatectic melts at UHT conditions are less dry than commonly assumed, with relevant
600 implications for the viscosity of anatectic melts. The production of low density rhyolitic melts
601 in the lower crust may induce large-scale weakening (Rosenberg and Handy, 2005) that
602 ultimately may lead to the lateral flow of mountain belts (Jamieson et al. 2011) or to the
603 formation of domes and crustal diapirs (e.g. Gerya, Perchuk, Maresch, & Willner, 2004;
604 Teyssier & Whitney, 2002). The latter mechanism can be an effective geological process that
605 may trigger the exhumation of high-grade metamorphic rocks. Moreover, the potassic to
606 ultrapotassic composition of the analysed anatectic melts may indicate that crustal anatexis can
607 have a larger impact in the redistribution of heat-producing elements (such as K₂O), with
608 important implications for the thermal structure of the continental crust.

609 This study shows that anatectic melt inclusions provide a fundamental insight into the
610 comprehension of the melting regime in UHT terranes and the chemical differentiation of the
611 lower crust.

612

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625

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	Type A granulites		Type C granulites	
	BRE-core	BRE-rim (glassy)	GRAG-core	GRAG-rim
No. analyses by EPMA ^a / NanoSIMS	6 / 6	11	21 / 3	6 / 7
Wt%				
SiO ₂	70.08 (2.51)	75.33 (1.26)	74.90 (1.00)	71.93 (1.54)
TiO ₂	0.05 (0.04)	0.00 (0.01)	0.05 (0.04)	0.06 (0.07)
Al ₂ O ₃	14.45 (0.88)	12.87 (0.64)	11.75 (0.43)	13.29 (1.19)
FeO	1.67 (0.32)	0.91 (0.05)	1.52 (0.14)	1.93 (0.36)
MnO	0.05 (0.02)	0.05 (0.02)	0.02 (0.02)	0.08 (0.04)
MgO	0.54 (0.24)	0.10 (0.08)	0.71 (0.10)	0.53 (0.23)
CaO	0.94 (0.55)	0.05 (0.02)	0.51 (0.15)	0.55 (0.19)
Na ₂ O	1.96 (0.54)	1.66 (0.38)	0.73 (0.27)	0.72 (0.39)
K ₂ O	7.31 (1.43)	8.67 (0.82)	6.96 (0.48)	7.95 (1.88)
P ₂ O ₅	0.20 (0.18)	0.04 (0.03)	0.12 (0.06)	0.17 (0.15)
Cl	0.04 (0.03)	0.01 (0.00)	0.06 (0.02)	0.12 (0.04)
Total	97.31 (2.33)	99.70 (0.31)	97.33 (0.79)	97.33 (1.57)
H ₂ O by NanoSIMS [wt%]	6.20 (1.25)	-	7.05 (3.00)	4.35 (0.92)
CO ₂ by NanoSIMS [ppm]	779 (326)	-	1113 (843)	940 (407)
ASI	1.14 (0.13)	1.06 (0.03)	1.24 (0.07)	1.27 (0.19)
K#	71 (8)	77 (5)	87 (4)	87 (7)
Mg#	35 (8)	13 (8)	44 (4)	32 (11)
CIPW norm				
Quartz	27.04	31.56	40.98	34.15
Corundum	2.10	0.76	2.39	2.89
Orthoclase	43.23	51.23	41.13	46.98
Albite	16.59	14.02	6.16	6.13
Anorthite	3.31	0.00	1.73	1.65
Hypersthene	4.43	2.01	4.51	4.91
Ilmenite	0.10	0.01	0.09	0.11
Apatite	0.47	0.10	0.29	0.39

^a EPMA = electron probe micro-analyzer

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989 **TABLE 1** Average compositions of remelted and preserved glassy MI. Numbers in parentheses
 990 refer to the standard error of the mean expressed as 2σ . ASI = molar
 991 $[Al_2O_3/(CaO+Na_2O+K_2O)]$, K# = molar $[K_2O/(K_2O+Na_2O)] \times 100$; Mg# = molar
 992 $[MgO/(MgO+FeO)] \times 100$.

Analysis	Sample	Approx. size	Homogeneity	H ₂ O [wt%]	1 σ error	CO ₂ [ppm]	1 σ error
<i>Type A granulites</i>							
C_MI1_Gr1	BRE17-B13 core	<10 μ m	Glass	5.52	0.04	460	20
C_MI3_Gr3_1	BRE17-B13 core	$\geq 20 \mu$ m	Glass + bubble	8.27	0.27	1164	33
C_MI3_Gr3_2	BRE17-B13 core	$\geq 20 \mu$ m	Glass + bubble	7.34	0.23	1064	31
C_MI16_Gr4	BRE17-B13 core	$\geq 20 \mu$ m	Glass + 10% crystals + bubble	6.88	0.21	1194	34
E_MI6_Gr2_1	BRE17-D2 core	<10 μ m	Glass	4.41	0.04	408	17
E_MI6_Gr2_2	BRE17-D2 core	<10 μ m	Glass	4.76	0.04	387	17
OG16_MI2_Gr1	BRE17-B13 rim	<10 μ m	Glass + 3% crystals	4.68	0.04	1153	32
<i>Type C granulites</i>							
A_MI1_Gr1	GRAG-P3 core	<10 μ m	Glass	8.75	0.07	1738	41
A_MI3_Gr2	GRAG-P3 core	<10 μ m	Glass + bubble	8.34	0.06	1291	33
A_MI2_Gr2	GRAG-P3 core	<10 μ m	Glass + 3% crystals	4.06	0.04	310	17
F_MI12_Gr1	GRAG-P2 Gr1 rim	10 - 20 μ m	Glass + 3% crystals	4.51	0.04	160	15
F_MI1_Gr4	GRAG-P2 Gr1 rim	<10 μ m	Glass + 2% crystals	2.88	0.04	1569	34
OG18_MI1_Gr4	GRAG-P3 rim	<10 μ m	Glass	3.11	0.04	1149	28
OG18_MI3_Gr4	GRAG-P3 rim	<10 μ m	Glass	3.44	0.04	1064	32
OG18_MI6_Gr1	GRAG-P3 rim	<10 μ m	Glass + 1% crystals	5.18	0.04	221	15
OG18_MI4_Gr2_1	GRAG-P3 rim	10 - 20 μ m	Glass	5.94	0.04	1287	31
OG18_MI4_Gr2_2	GRAG-P3 rim	10 - 20 μ m	Glass	5.41	0.04	1132	27

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994 **TABLE 2** H₂O and CO₂ contents of re-homogenized MI determined by NanoSIMS.

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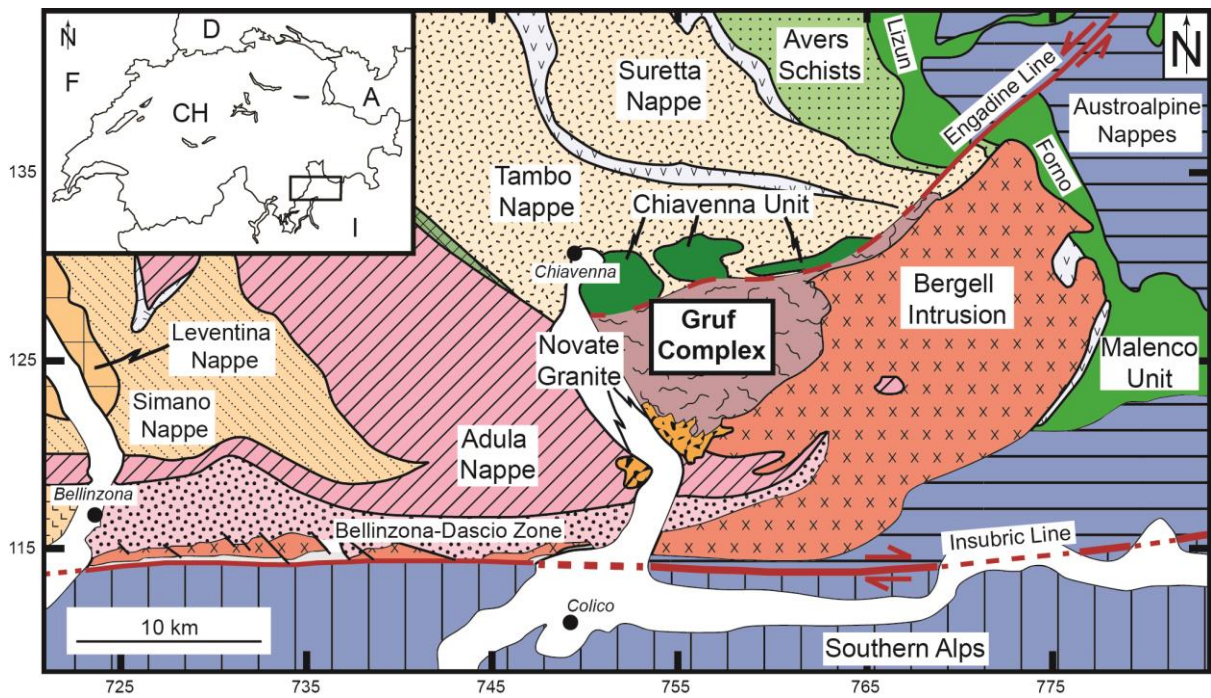
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1 **FIGURES TEXT**



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3 **FIGURE 1** Tectonic map of the Central Alps modified after **Trommsdorff and Nievergelt**
4 **(1983)**. Coordinates are given according to the Swiss geographical grid (units in kilometres).

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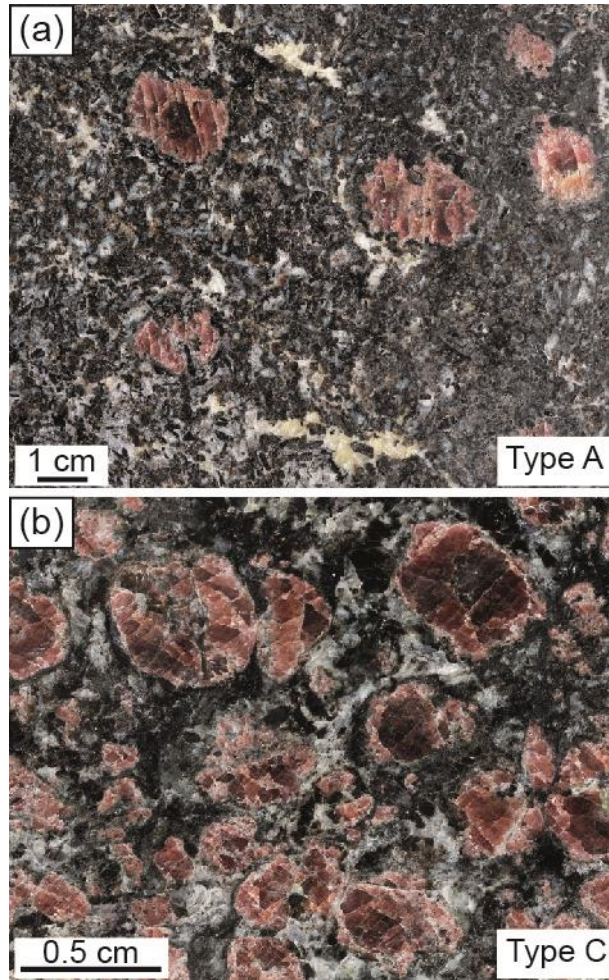
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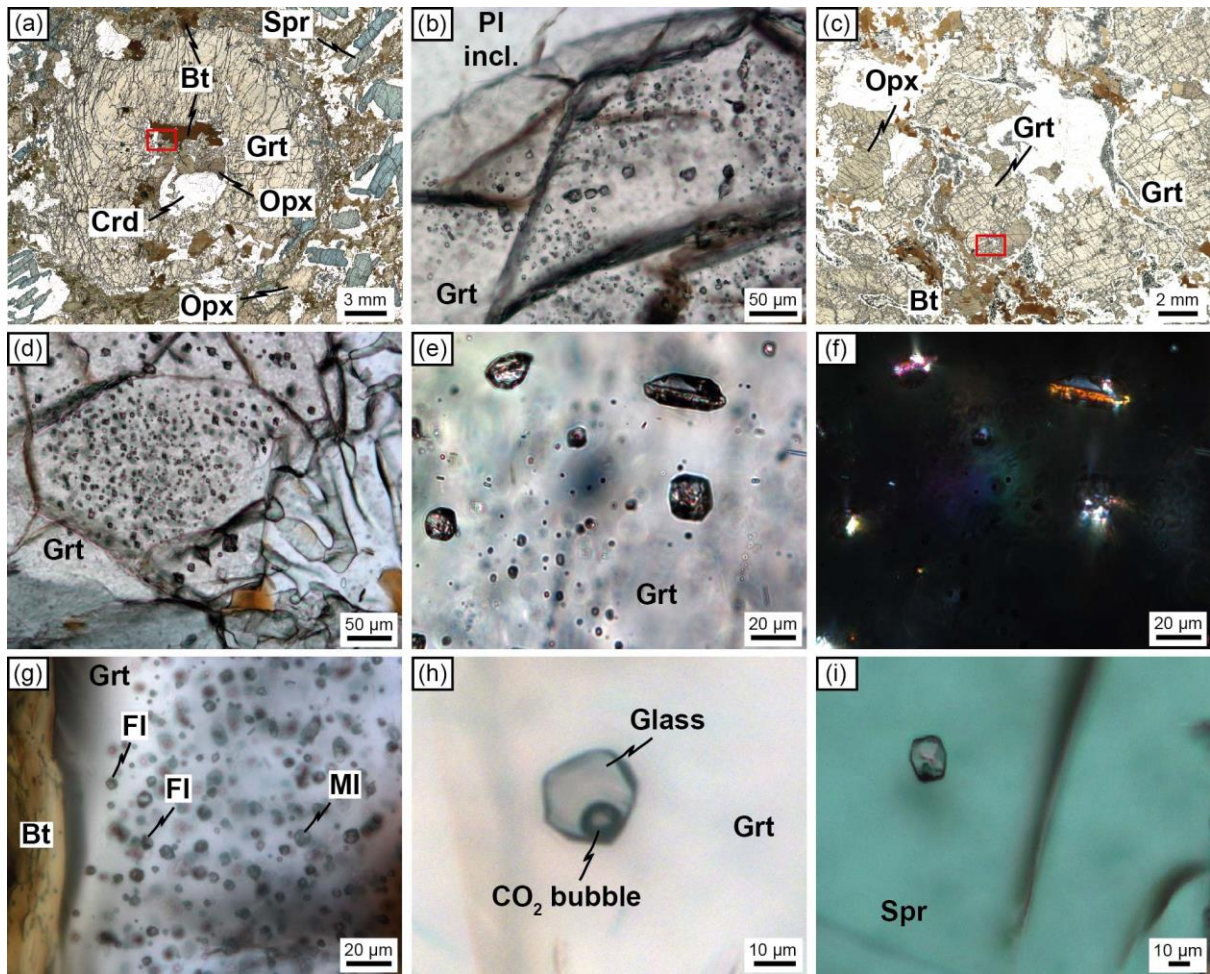
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17 **FIGURE 2** UHT granulites from the Gruf Complex. (a) Massive sapphirine-orthopyroxene-
18 cordierite-garnet granulite (Type A) and (b) massive orthopyroxene-cordierite-garnet granulite
19 (Type C).

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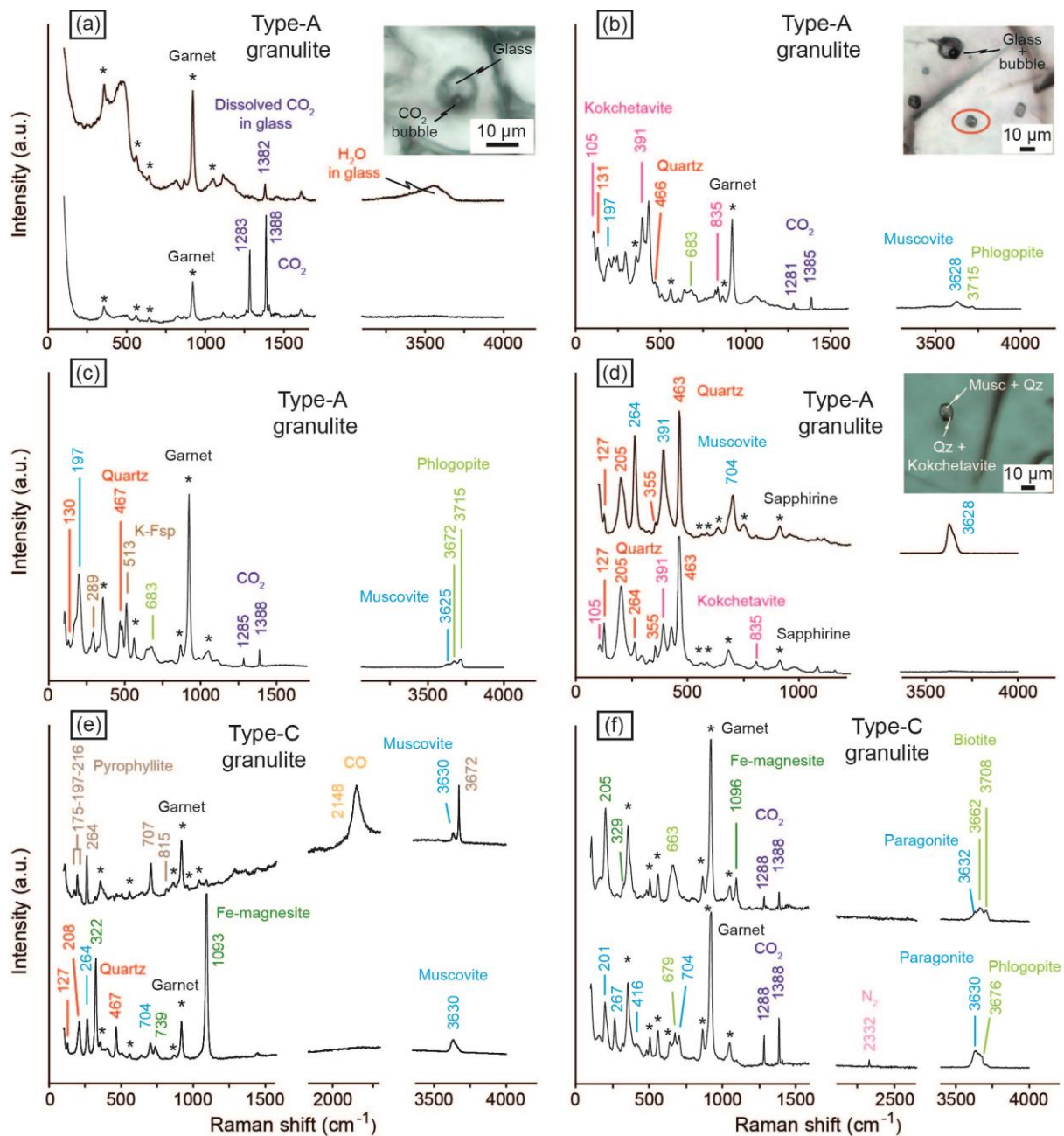
22 **FIGURE 3** Photomicrographs of the granulites from the Gruf Complex. (a) Garnet
 23 porphyroblast from a Type A granulite showing a cluster of MI near to the core of the crystal
 24 (red box). (b) Zoom of the same cluster of MI as in (a). (c) Garnet porphyroblasts from a Type
 25 C granulite. The red box at the rim of the garnet indicate a cluster of MI. (d) Zoom of the same
 26 cluster of MI as in (c). (d) Plane-polarized light photomicrograph of MI in garnet. (e) Same as
 27 (d) under crossed-polarized light, showing multiple birefringent phases within the inclusions.
 28 (g) Fluid inclusions (FI) and MI within a cluster. (h) Glassy MI containing a CO₂ shrinkage
 29 bubble. (i) MI in a sapphirine porphyroblast from a Type A granulite.

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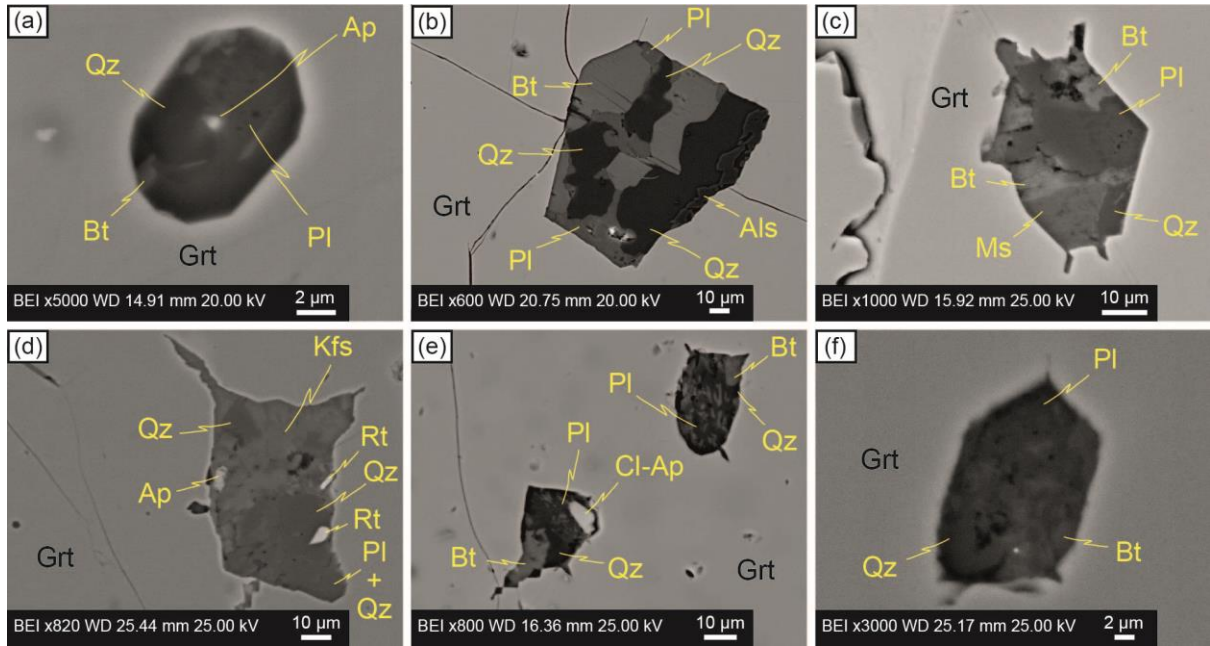


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35 **FIGURE 4** Raman spectra of nanogranitoids and glassy MI. (a) Glassy MI with a shrinkage
 36 bubble containing CO₂. (b) Analysis of a nanogranitoid inclusion (red circle) close to a glassy
 37 MI showing a shrinkage bubble. (c) Nanogranitoid with a CO₂ bubble. (d) Nanogranitoid in
 38 sapphirine. (e) Mixed inclusion (fluid + melt) showing CO together with phyllosilicates, quartz
 39 and Fe-magnesite. (f) Mixed inclusion with phyllosilicates, Fe-magnesite, CO₂ and N₂.

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43 **FIGURE 5** Backscattered images of nanogranitoids. Nanogranitoids from Type A (a-d) and
 44 from Type C (e-f) granulites are shown.

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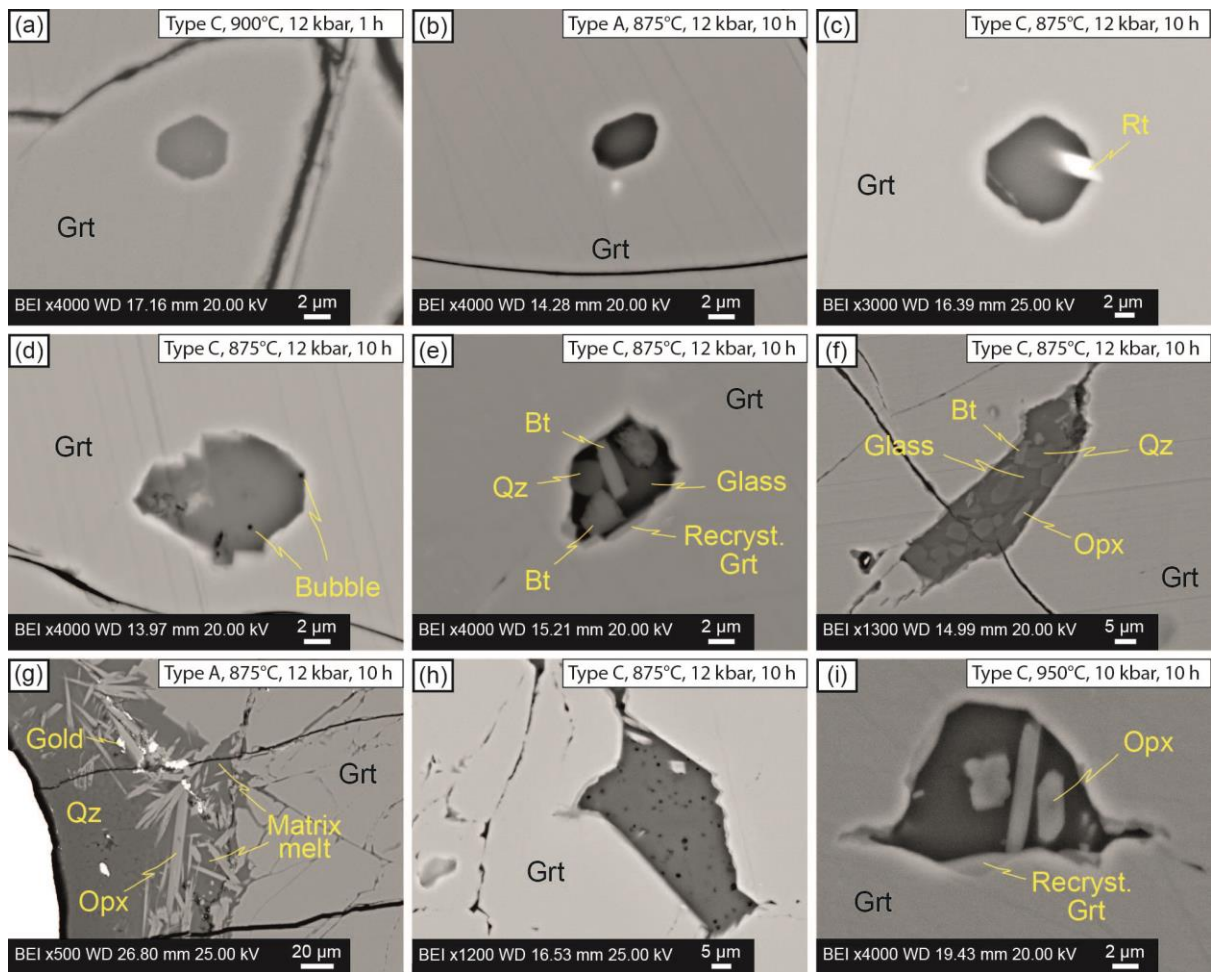
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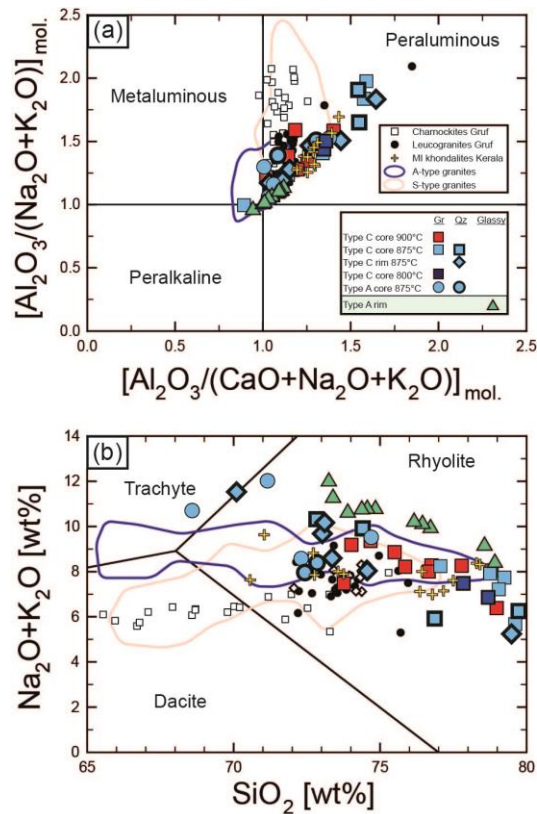
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60 **FIGURE 6** Backscattered images of experimentally remelted nanogranitoids. (a,b) Remelted
 61 MI with a well-developed negative shape. (c) Re-homogenized MI showing a trapped rutile
 62 needle, indenting the wall of the inclusion. (d) MI showing an irregular border and bubbles
 63 within the glass. (e,f) Partially remelted nanogranitoids, displaying daughter phases (biotite
 64 and quartz) and evidence of minor overheating (recrystallization of garnet and peritectic
 65 orthopyroxene). (g) Interaction of matrix melt with a garnet chip, forming peritectic
 66 orthopyroxene needles. (h) Melt pocket of infiltrated matrix melt within the garnet chip. The
 67 glass displays abundant bubbles. (i) MI from an experiment at 950 °C showing strong evidence
 68 of overheating (formation of peritectic orthopyroxene and recrystallization of new garnet at the
 69 wall of the inclusion).

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72 **FIGURE 7** Chemical classification of remelted and glassy MI from the Gruf Complex

73 (normalized to 100% on anhydrous basis). Reported are MI analyses of re-melting

74 experiments between 800 and 900 °C with graphite-bearing (Gr) and quartz-bearing (Qz)

75 capsules. (a) Aluminum saturation index vs. alkalinity index. (b) TAS diagram. Analyses for

76 charnockites and leucogranites from the Gruf Complex are from [Galli et al. \(2013\)](#), whereas

77 MI analyses from the Kerala Khondalite Belt are from [Cesare et al. \(2009\)](#) and [Ferrero et al.](#)

78 [\(2012\)](#). Data for S-type granites ([Chappel & White, 1992](#); [Jung, Hoernes, & Mezger, 2000](#);

79 [Kalsbeek, Jepsen, & Jones, 2001](#); [Tulloch, Ramezani, Kimbrough, Faure, & Allibone, 2009](#);

80 [Visonà & Lombardo, 2002](#)) and for A-type granites ([Castro et al., 2012](#); [de Almeida, de](#)

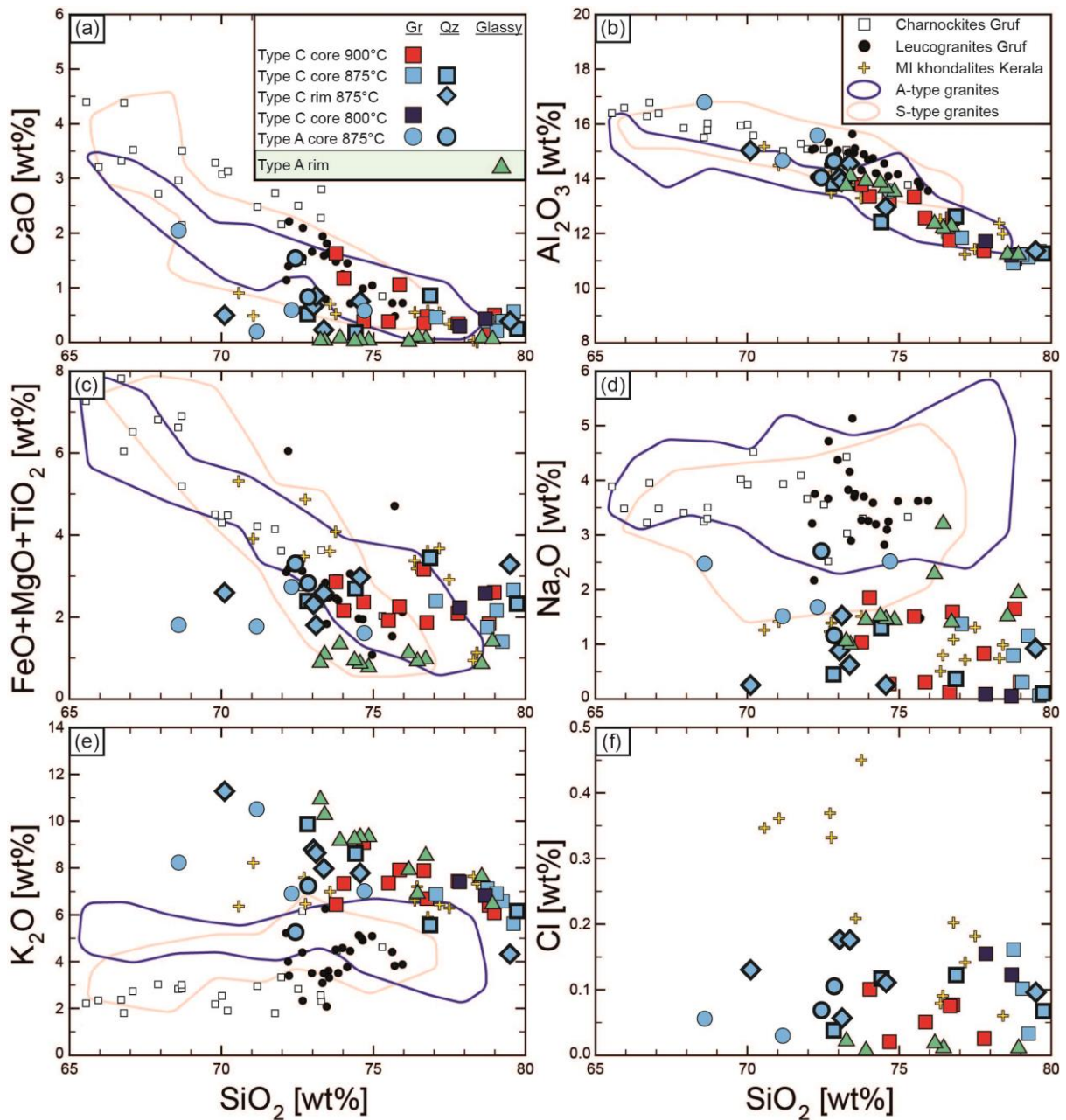
81 [Pinho Guimarães, & da Silva Filho, 2002](#); [Floribal, de Fátima Bitencourt, Nardi, &](#)

82 [Conceição, 2009](#); [Ghani et al., 2014](#); [Goodenough, Upton, & Ellam, 2000](#); [King, White,](#)

83 [Chappell, & Allen, 1997](#); [King, Chappell, Allen, & White, 2001](#); [McDermott, Harris, &](#)

84 [Hawkesworth, 2000](#); [Whalen, Currie, & Chappell, 1987](#); [Zhang et al., 2007](#)) are also reported

85 for comparison.



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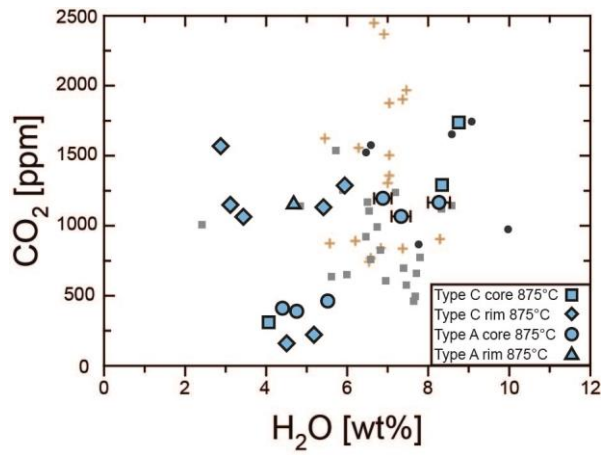
87 **FIGURE 8** Chemical composition of glasses from experimentally remelted and natural glassy
 88 inclusions (normalized to 100% on anhydrous basis). Reference analyses are the same as in
 89 Figure 7.

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96 **FIGURE 9** H₂O and CO₂ content measured in remelted nanogranitoid inclusions. Error bars

97 are displayed only when bigger than the symbol. Data for MI from the Ivrea Zone (Carvalho

98 et al. 2019) are also reported for comparison (black dots = upper amphibolite facies, grey

99 squares = transition zone, brown crosses = granulite facies).

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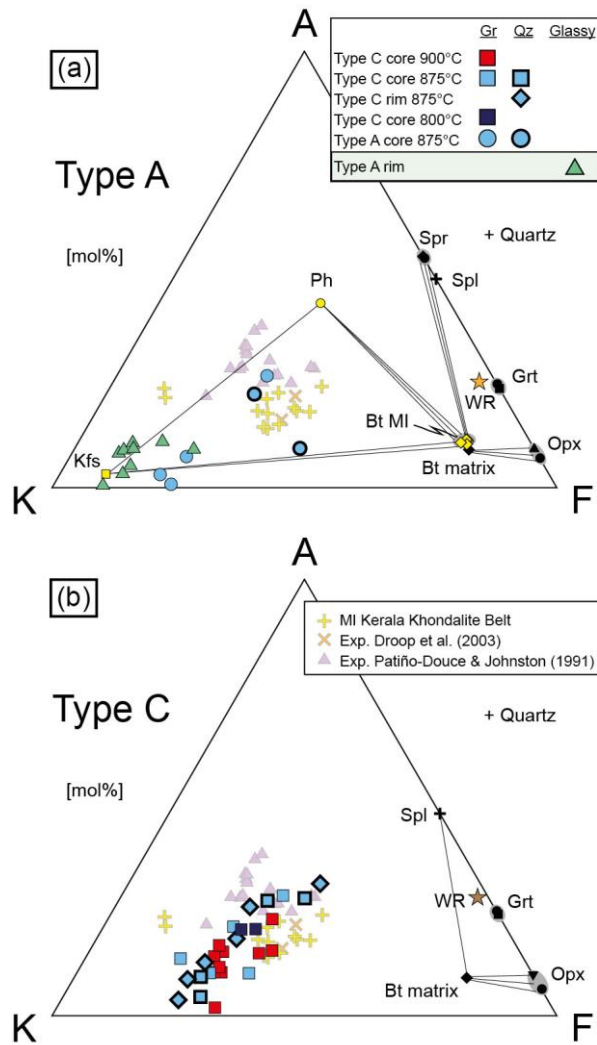
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112 **FIGURE 10** AKF diagram (after Vielzeuf & Holloway, 1988) projected from quartz for MI

113 and bulk rocks. (a) Type A granulite. (b) Type C granulite. Also reported for comparison are:

114 melt inclusions from the Kerala Khondalite Belt (Cesare et al. 2009; Ferrero et al. 2012),

115 experimental glasses (900 °C, 5 kbar) of Droop et al. (2003) and experimental glasses (875-

116 1075 °C, 7-13 kbar) of Patiño-Douce & Johnston (1991).

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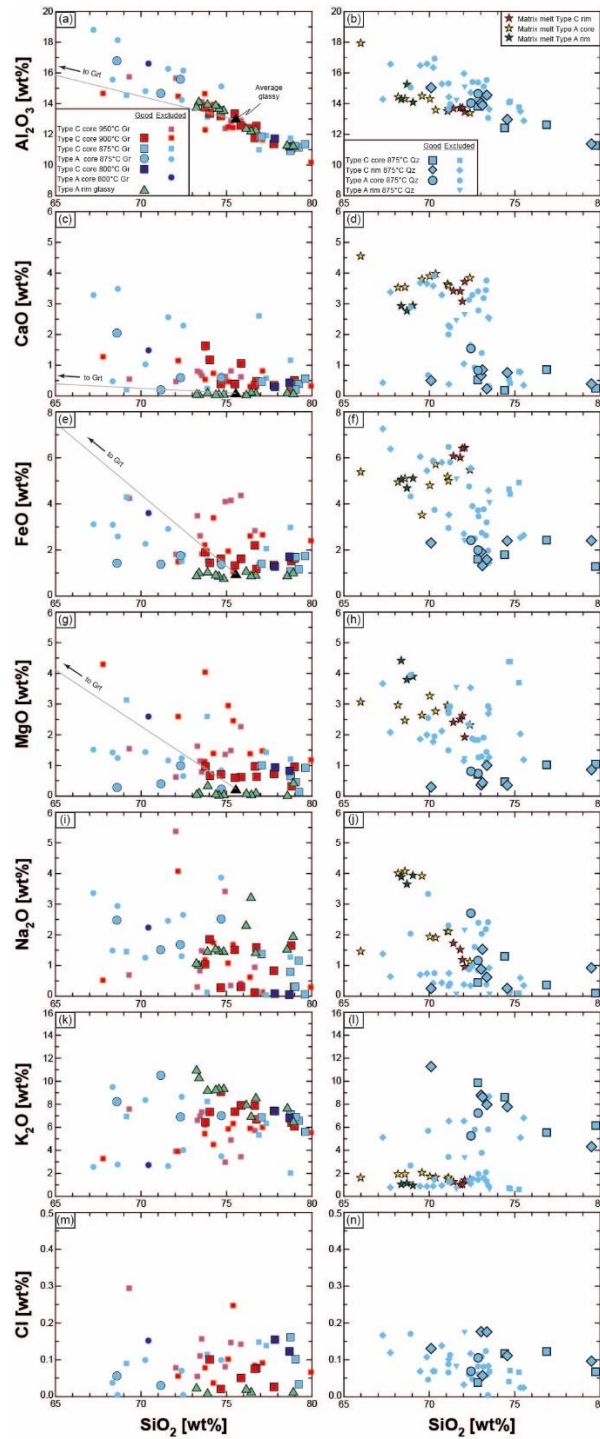
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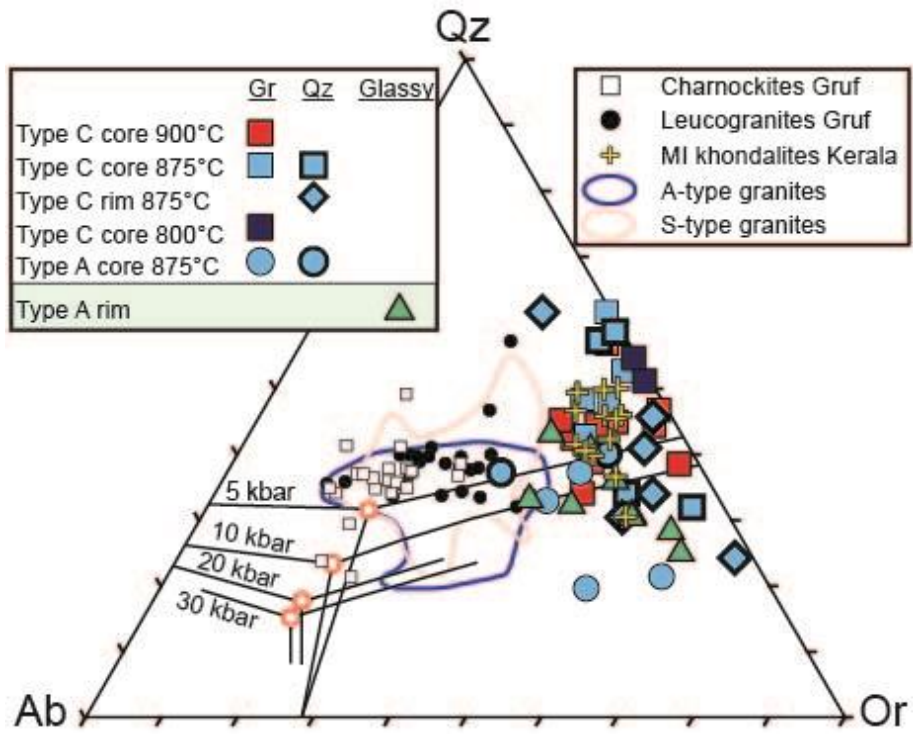


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125 **FIGURE S1** Harker diagrams showing the complete set of MI analyses. Small symbols
 126 represent disregarded analyses. The line displayed for Al₂O₃, FeO and and MgO represents
 127 the tie-line between the host garnet (Type A granulite) and the average composition of the
 128 glassy inclusions (black triangle).

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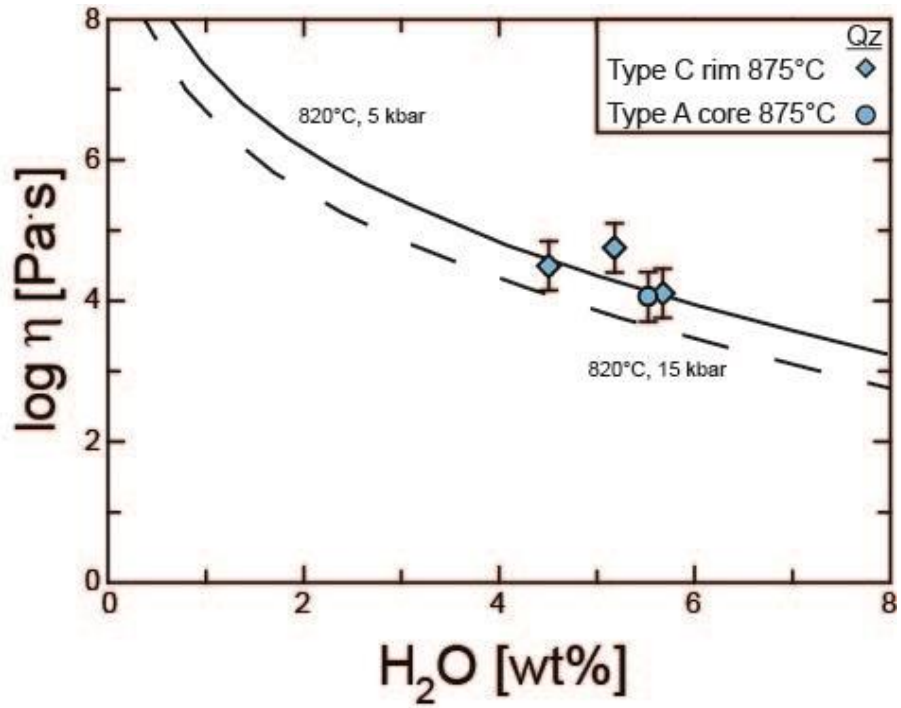
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133 **FIGURE S2** CIPW Qz-Ab-Or diagram showing normative compositions for the analysed MI
134 and other reference analyses from the literature (see text).

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139 **FIGURE S3** Viscosities of rhyolitic melts vs. H₂O content (modified after Ardia et al.,
 140 2008). Viscosities for MI are calculated according to Giordano et al. (2008).

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